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## (54) THERMOPLASTIC ELASTOMER COMPOSITION

## (57)Abstract:

PROBLEM TO BE SOLVED: To provide a new thermoplastic elastomer composition which has flexibility and has excellent formability, rubbery characteristics, mechanical strength, compression permanent set, and a vibration-damping property.

SOLUTION: This thermoplastic elastomer composition comprises (A) an isobutylene block copolymer comprising a polymer block consisting mainly of isobutylene and a polymer block consisting mainly of an aromatic vinyl compound and (B) a modified isobutylene block copolymer which comprises the block copolymer comprising isobutylene and an aromatic vinyl compound and having alkenyl groups at the terminals.

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### CLAIMS

[Claim(s)]
[Claim(s)]
[Claim 1] The thermoplastic-elastomer constituent which comes to blend the denaturation isobutylene system block copolymer (B) which are an isobutylene system block copolymer (A) containing the polymer block which makes a subject the polymer block and aromatic series vinyl system compound which make an isobutylene a subject, and a block copolymer containing the polymer block which makes a subject the polymer block and aromatic series vinyl system compound which make an isobutylene a subject, and has an afternyl radical at the end [claim 2] A denaturation isobutylene system block copolymer (B) is a thermoplastic-elastomer constituent according to claim 1 with which an allyl group is introduced into an end by the substitution reaction of an allyl compound trimethyl silane and chlorine.
[Claim 3] The thermoplastic-elastomer constituent according to claim 1 or 2 with which a

according to claim 1 with which an allyl group is introduced into an end by the substitution reaction of an allyl compound trimethyl silane and chlorine.

(Claim 3) The thermoplastic-elastomer constituent according to claim 1 or 2 with which a thermoplastic-elastomer constituent active of melting kneading of an isobutylene system block copolymer (A) and a denaturation isobutylene system block copolymer (A) and a denaturation isobutylene system block copolymer (B).

(Claim 4) The thermoplastic-elastomer constituent according to claim 1 or 2 characterized by a denaturation isobutylene system block copolymer (B) constructing a bridge beforehand before being mixed with an isobutylene system block copolymer (B).

(Claim 5) from the polymer block (a) with which the block which constitutes an isobutylene system block copolymer (B).

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(Claim 6) The thermoplastic-t and the polymer block (b) which makes an aromatic series vinyl system compound a subject — becoming — (b)—(a)—the thermoplastic-elastomer constituent according to claim 1 to 5 which furthermore contains a plasticizer (C).

(Claim 7) The thermoplastic-elastomer constituent according to claim 6 with which a plasticizer (C) is chosen from paraffin series and naphthene straight mineral oil and which is a kind at least.

(Claim 8) The thermoplastic-elastomer constituent according to claim 1 to 7 which furthermore contains a cross linking agent (D).

(Claim 8) The thermoplastic-elastomer constituent according to claim 8 whose cross linking agent (D) and is the block copolymer (B) is a thermoplastic-elastomer constituent according to claim 1 to 5 characterized by carrying out 10–300 weight section content of the denaturation isobutylene system block copolymer (B) with which the alternyl radical was introduced into the end to the is

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### DETAILED DESCRIPTION

### [Detailed Description of the Invention]

[Field of the Invention] This invention is rich in flexibility and relates to the new thermoplasticelastomer constituent excellent in fabrication nature, a rubber transparency, and a compression set property. transpa [0002]

[0002] [Description of the Prior Art] Conventionally, what blended the cross linking agent, the reinforcing agent, etc. with rubber, such as natural rubber or synthetic rubber, and constructed the bridge under elevated-temperature high pressure as polymeric materials which have elasticity is used widely. However, with such rubber, the stroke which performs bridge formation and shaping over long duration under elevated-temperature high pressure is required, and it is inferior to workability. Moreover, since the rubber which constructed the bridge does not show thermoplasticity, generally recycle shaping is impossible for it like thermoplastics. Therefore, the thermoplasticity, generally recycle shaping is impossible for it like thermoplastics using general-purpose mething forming technique, such as heat press forming, injection molding, and extrusion molding, is developed variously in recent years. Moreover, the elasticity vinyl chloride compound is used widely as an ingredient which has flexibility. Although this is used for various applications as a flexible ingredient at the room temperature, the alternative with other ingredients is demanded from the demand of the formation of devirnyl chloride in recent years. The thermoplastic elastomer constituent is used as alternate material for this. The polymer of various formats, such as current, an olefin system, an urethane system, an ester system, a styrene system, and a vinyl chloride system, is developed and marketed by such thermoplastic elastomer.

styrene system, and a vmy crieorios system, is developed and marketed by such derimpleasure elastomer.

[0003] A styrene thermoplastic elastomer is rich in flexibility, and excellent in good rubber elasticity with ordinary temperature. [among these ] As a styrene thermoplastic elastomer, the styrene-butadiene-styrene block copolymer (SES), the styrene-styrene styrene block copolymer (SES), a styrene-ethylene butylene-styrene block copolymer (SES), a styrene-ethylene propylene-styrene block copolymer (SES) that hydrogensted them, etc. are developed. However, these block copolymers had the inadequate compression set property.

[0004] The isobutylene system block copolymer which contains the polymer block which makes an isobutylene a subject, and the polymer block which makes an aromatic series vinyl system compound a subject as thermoplastic elastomer which was rich in flexibility, was excellent in good rubber elasticity on the other hand with ordinary temperature, and was further excellent in gas barrier nature and sealing performance is known. However, this isobutylene system block copolymer also had a problem in the pressurization reduction of area at the time of heating (compression set), or the rubber elasticity at the time of an elevated temperature.

[0005] Moreover, the thermoplastic polymerization body composition object which consists of the isobutylene system block copolymer and the bridge formation object of rubber containing the polymer block which makes an isobutylene a subject is known (re-official announcement patent WO 98/14518). Although the compression set property has been improved and the compression set has been improved, this constituent was opaque and its degree-of-hardness-mechanical

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styrene, an indene, etc. are mentioned. Also in the above-mentioned compound, styrene, alpha methyl styrene, p-methyl styrene, and an indene are desirable from the balance of cost, physical properties, and productivity, and you may also choose two or more sorts from them. [0014] It is related with the rate of the polymer block (b) which makes a subject the polymer block (c) which makes a subject the isobutylene in an isobutylene system block copolymer (A), and an aromatic series vinyl compound. Although there is especially no limit, the polymer block (a) which makes an sisobutylene a subject from the balance of physical properties and workability of the polymer block (b) which makes an aromatic series vinyl compound a subject is 5 - 80 weight section. It is desirable that the polymer block (b) which makes an aromatic series vinyl compound a subject is 5 - 80 weight section and an aromatic series vinyl compound is especially 10 - 40 weight section. [0015] Moreover, the structure which consists of at least one of the polymer blocks (a) which make an isobutylene a subject from the point of the physical properties of the constituent obtained and workability, and at least two of the polymer blocks (b) which make an aromatic series vinyl system compound a subject as desirable structure of the isobutylene system block copolymer (A) of this invention is desirable, the above — structure — \*\*\*\*\* — especially — a finite— there is nothing — although — for example — (— b —) — (— a —) — — a unit — a repeat — having — a triblock — a copolymer — |— (— b —) — (— a —) — a unit — a repeat — having — a triblock — a copolymer — and — (— b —) — the jib which consists of (a) — at least one sort chosen from the star-like polymer which uses a lock copolymer as an arm can be used, furthermore, the polymer which makes an isobutylene a subject into an isobutylene system block copolymer (A) in addition to the above—mentioned structure, the polymer which makes an aromatic series vinyl system compound a subject, and (a)— the jib which consis a subject becomes 50% of the weight or more from the point of physical prop

a subject becomes 50% of the weight or more from the point of physical properties and workability.

[0016] Although there is especially no limit also in the weight average molecular weight of an isobutylene system block copolymer (A), 30,000–500,000 are desirable, and 40,000 to especially 400,000 is good \*\*\*\*. The fall of a moldability etc. is large when a mechanical property etc. is not fully discovered when weight average molecular weight is less than 30,000, and exceeding 500,000. It is a block copolymer containing the polymer block which makes a subject the polymer block and aromatic series viryl system compound as used in the field of this invention which make an isobutylene a subject, and the denaturation isobutylene system block copolymer (B) which has an alkenyl radical at the end is the object which denaturalized the end of the copolymer shown with the isobutylene system block copolymer (A) by the alkenyl radical. Therefore, the polymer block of the component which constitutes the above-mentioned isobutylene system block copolymer (A) which makes a subject the polymer block and aromatic series viryl system compound which make an isobutylene a subject may be the same in molecular weight etc. respectively, and may differ. However, the polymer blocks which make an isobutylene a subject may be the same in compound a subject may be the same configurations, respectively, and you may differ. [0017] Although there is especially no limit also in the weight average molecular weight of a denaturation isobutylene system block copolymer (B), 1,000-500,000 are desirable, and 2,000 to especially 100,000 is good \*\*\*\*. The fall of a moldability etc. is large when a mechanical property etc. is not fully discovered when weight average molecular weight is less than 1,000, and exceeding 500,000. ting 500,000

exceeding 500,000.
[0018] It will not be restricted especially if the alkenyl radical of this invention is a radical including the carbon-carbon double bond which has activity to the crosslinking reaction of the (B) component for attaining the purpose of this invention. As an example, ring type unsaturated hydrocarbon radicals, such as a lighthic series partial saturation hydrocarbon groups, such as a vinyl group, an allyl group, a methylvinyl radical, a propenyl radical, a butenyl group, a pentenyl

strength balance was also inadequate. [00006]

[0006]
[Problem(a) to be Solved by the Invention] In view of the technical problem of the above—mentioned conventional technique, the purpose of this invention is rich in flexibility, and is to offer the fabrication nature, rubber-property, mechanical-strength, and thermoplastic-elastor constituent which was transparent and was excellent in the compression set property.

[Means for Solving the Problem] That is, this inventions are an isobutylene system block copolymer (A) containing the polymer block which makes a subject the polymer block and aromatic series viryl system compound which make an isobutylene a subject, and a block copolymer containing the polymer block which makes a subject the polymer block and aromatic series viryl system compound which make an isobutylene a subject, and are thermoplastic series viryl system compound which make an isobutylene a subject, and are thermoplastic-elastomer constituents which come to blend the denaturation isobutylene system block copolymer (B) which has an alkemyl radical at the end. Said denaturation isobutylene system block copolymer (B) has that desirable by which the allyl group was introduced into the end by the substitution reaction of an allyl compound trimethyl silane and chlorine. Moreover, as thermoplastic-elastomer constituent, a bridge can be dynamically constructed in a denaturation isobutylene system block copolymer (B) at the time of melting kneading of an isobutylene system block copolymer (A) and a denaturation isobutylene system block copolymer (B), and a denaturation inch thems averame block component (B) can also construct a bridge beforehand.

block copolymer (A) and a densturation isobutylene system block copolymer (B), and a densturation isobutylene system block copolymer (B), and a densturation isobutylene system block copolymer (B) can also construct a bridge beforehand, before being mixed with an isobutylene system block copolymer (A).

[0008] from the polymer block (a) with which the block which constitutes said isobutylene system block copolymer (A) and/or denaturation isobutylene system block copolymer (B) which were carried out as structure of a block copolymer makes an isobutylene a subject, and the polymer block (b) which makes an aromatic series viryl system compound a subject — becoming — (b)–(a)— it is desirable that it is the triblock copolymer in which the structure of (b) is shown.

[0009] As a thermoplastic—elastomer constituent, a plasticizer (C) can be contained further and a cross finding agent (D) can also be contained further.

[010]

[Embodiment of the Invention] The thermoplastic-elastomer constituents of this invention are an isobutylene system block copolymer (A) containing the polymer block which makes a subject the polymer block and aromatic series viryl system compound which make an isobutylene a subject, and a block copolymer containing the polymer block which makes a subject the polymer block and aromatic series viryl system compound which make an isobutylene a subject, and are thermoplastic-elastomer constituents which come to blend the denaturation isobutylene system

and aromatic series why system compounts which make an associty-real subject, and are thermoplastic-elastomer constituents which come to blend the denaturation isobutylene system block copolymer (B) which has an alkernyl radical at the end. [0011] The polymer block which makes a subject the isobutylene of the isobutylene occupies 90 % of the weight or more more preferably 70% of the weight or more 50% of the weight or more. Although monomers other than an isobutylene under polymer block which makes an isobutylene a subject will not be limited especially if they are monomer components in which cationic polymerization is possible, they can illustrate monomers, such as aromatic series vinyl, alphatic series olefins, dienes, vinyl ether, and beta-pinene. These may be used independently, and two or more sorts may be combined and they may be used. [0012] The polymer block which makes a subject the aromatic series vinyl system compound of an isobutylene system block copolymer (A) means the block with which an aromatic series vinyl system compound of capital propound occupies 90 % of the weight or more softs of the weight or more 50% of the weight or more. Although there will be especially no limit if it is the monomer in which cationic polymerization is possible as monomers other than the aromatic series vinyl compound under polymer block which makes an aromatic series vinyl system compound a subject, monomers, such as alighatic series olefins, dienes, vinyl ether, and beta-pinene, can be illustrated.

Stagets, incomments and illustrated.
[0013] As an aromatic series viryl system compound, styrene, alpha methyl styrene, beta-methyl styrene, p-methyl styrene, t-butyl styrene, monochlorostyrene, dichloro styrene, methoxy

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radical, and a hexenyl radical, a cyclo propenyl radical, a cyclo butenyl group, a cyclo pentenyl

radical, and a hexenyl radical, a cyclo propenyl radical, a cyclo butenyl group, a cyclo pentenyl radical, and a cyclohexeryl group, can be mentioned.

[0019] The approach of making the compound which has a partial saturation radical react to the polymer which has functional groups, such as a hydroxyl group which is indicated by JP.3-152164.A and JP.7-304909.A as the introductory approach of the alkenyl radical to the end of the isobutylene system block copolymer of this invention, and introducing a partial saturation radical into a polymer is mentioned. Moreover, the method of performing the further aforementioned alkenyl radical into a holymer is mentioned. Moreover, the method of performing the further aforementioned alkenyl radical intallation reaction in order to introduce a partial saturation radical into the polymer which has a halogen atom, after performing the Friedel Crafts reaction with the approach and the various phenols which perform a substitution reaction with the approach of performing an Friedel Crafts reaction with alkenyl phenyl ether, a bottom allyl compound of Lewis acid existence trimethyl silane, etc. and introducing a hydroxyl group etc. is mentioned. It is also possible to introduce a partial saturation radical at the time of the polymerization of a monomer as furthermore indicated by U.S. Pat. No. 4316973, JP,63-103005.A, and JP.4-288309.A. That by which the allyl group was especially introduced into the end by the substitution reaction with an allyl compound trimethyl silane is desirable. Moreover, as for the alkenyl radical permuted by the end, it is desirable to exist in at least 0.2 ends per molecule. When fewer than this, the constituent excellent in the compression set is not obtained.

[0020] the constituent which constructed the bridge beforehand in the denaturation isobutyles system block copolymer (B) with which the alkernyl radical was introduced into the end or the thermoplastic-elastomer constituent which consists of an isobutylene system block copolyme (A) and a denaturation isobutylene system block copolymer (B) with which the alkernyl radical was introduced into the end would not construct a bridge dynamically at the time of melting kneading, and carried out mehting mixing of the isobutylene system block copolymer (A) from the system block copolymer (B) with the system block copolymer (B) with the system block copolymer (B) with which the system block copolymer (B) with which the silver in the system block copolymer (B) with which the silver in the system block copolymer (B) with which the silver in the system block copolymer (B) with which the silver in the system block copolymer (B) with which the silver in the system block copolymer (B) with which the silver in the system block copolymer (B) with which the silver in the system block copolymer (B) with which the silver in the system block copolymer (B) with which the silver in the system block copolymer (B) with which the system block copolymer (B) with which the system is system block copolymer (B) with which the system is system block copolymer (B) with which the system is system in the system is system in the system is system in the system in the system

desirable.

[0021] In the rack abutment formed here, the object with which (B) constructed the bridge independently, and the object with which (A) and (B) were contained in the rack abutment, and constructed the bridge over coincidence is contained. It is desirable to form a bridge formation object by the (B) independent among these.

[0022] Although a well-known approach can be used as a means to construct a bridge in the isobutylene system block copolymer (B) with which the alkenyl radical was introduced into the end and there is especially no limit, radical crosslinking can also be performed, for example without using the heat bridge formation by heating, the bridge formation by the cross linking agent (D), or a cross linking agent.

[0023] It is desirable to use a hydrosilyl radical content compound as a cross linking agent (D) for obtaining the bridge formation object of the isobutylene system block copolymer (B) with which the alkenyl radical was introduced into the end of this invention. As a hydrosilyl radical content compound, there is especially no limit and it can use various kinds of things. Namely, chain-like polysilosane expressed with a general formula (I) or (II):

R13SIO-[CS(R1)2O]a-[CS(R2)]

R13SiO-[Si(R1)2O]=-[Si(H)(R2)O]A-[Si(R2) (R3)O]B-SiR13 (I) HR12SiO-[Si(R1)2O]=-[Si(H)(R2)O]A-[Si(R2)(R3)O]B-SiR12H (II)

(As for R1 and R2, the akky group of carbon numbers 1=6 or a phenyl group, and R3 show the alkyl group or aralkyl radical of carbon numbers 1=10 among a formula.) a shows the integer with which OC=aC=100 and A fill 2C=AC=100, and B fills OC=BC=100. Cyclosiloxane expressed with a general formula (III):

[0024] [Formula 1]

(As for R4 and R5, the aByl group of carbon numbers 1–6 or a phenyl group, and R6 show the aByl group or araByl radical of carbon numbers 1–10 among a formula.) 0C=CC=8 and e express 2C=cC=10, and f expresses the integer of 0C=CC=8, and C fills 3 CC+c+CF=C0, etc. — a compound can be used. What compatibility with the (B) component is especially expressed with the following general formula (IV) to from the point of being good, among the compounds which have the further above-mentioned hydrosilyl radical (Si-H radical) is desirable. have th [0025]

[Formula 2]

CH<sub>3</sub> CH<sub>3</sub> CH<sub>3</sub> CH<sub>3</sub> CH<sub>3</sub>

(Among a formula, g and h are integers and are 2 <=g+h<=50, 2<=g, and 0<=h.) R7 may express a hydrogen atom or a methyl group, and R8 may have one or more rings by the hydrocarbon group of carbon numbers 2-20, is the integer of 0<=i<-5.

[0026] Although the denaturation isobutylene system block copolymer (B) and cross linking agent by which the alkenyl radical was introduced into the end are mixable at a rate of arbitration, it is desirable that the mole ratio of an alkenyl radical and a hydrosilyl radical is in the range of 0.2-5 from the field of hardenability, and it is still more desirable that it is especially 0.4-2.5, if a mole ratio becomes five or more, bridge formation is inadequate, if smaller than constituent profit \*\*\*e\*\* with reinforcement of enough, and 0.2, since an activity hydrosilyl radical remains in large quantities into a constituent, after bridge formation will be uniform, and a constituent with reinforcement will not be obtained.

[0027] Although the crosslinking reaction of a copolymer (B) and a cross linking agent (D) advances by mixing and heating two components, in order to advance a reaction more quickly, it can add a hydrosilylation catalyst. It is not limited especially as such a hydrosilylation catalyst, for example, radical initiators, such as organic peroxide and an azo compound, and a transition metal catalyst are mentioned.

[0028] It is not limited especially as a radical initiator. For example, G t-butyl peroxide, 2, the 5-dimethyl −2, 5-JI (t-butylperoxy) -hexpine, C, the 5-dimethyl −2, 5-JI (t-butylperoxy) -hexpine, 2, the 5-dimethyl −2, 5-JI (t-butylperoxy) -hexpine, peroxide, discyl peroxide like fauroyl peroxide, m-chloro benzoyl peroxide, m-chloro benzoyl peroxide, m-chloro benzoyl peroxide, m-chloro benzoyl peroxide, carbonic acid G 2-ethylhexyl J, 1, and 1-JI (t-butylperoxy) -peroxy dicarbonate [ like fauth JI carbonic acid diisopropyl, Peroxi dicarbonate [ like fauth JI carbonic acid disophorever, a complex with the thing which it is (0)-diaryl tetramethyl disiloxane complex are mentioned. As an example of catalysts other than a

If loadings exceed the 300 weight sections, a problem will arise in a fall and moldability of a

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If loadings exceed the 300 weight sections, a problem will arise in a fall and moldability of a mechanical strength. Moreover, the demand characteristics further doubled with each application at the constituent of this invention are accepted. In the range which does not spoil physical properties, a reinforcing agent and a bulking agent, for example, a styrene-butadiene-styrene block copolymer (SSS), Moreover, elastomers, such as a styrene-ethylene butylene-styrene block copolymer (SESS) which hydrogenated them, and a styrene-ethylene propylene-styrene block copolymer (SEPS), in addition to this, the antioxidant of a hindered phenol system or a hindered amine system, an utraviolet ray absorbent, light stabilizer, a pigment, a surfactant, a reaction retarder, a flame retarder, a bulking agent, a reinforcing agent, etc. can be blended suitably.

[0037] As most desirable constituent of the thermoplastic-elastomer constituent of this invention denaturation isobutylene system block-copolymer (B) 10-300 by which the alternyl radical was introduced into the end to the isobutylene system block-copolymer (A) 100 weight retarder, a bulking agent, a 'reinforcing agent, etc. can be blended suitably. (0037) As most desirable constituent of the thermoplastic-elastomer constituent of this invention denaturation isobutylene system block-copolymer (B) 10–300 by which the alkenyl radical was introduced into the end to the isobutylene system block-copolymer (A) 100 weight sections, and a pan an isobutylene — a system — a block copolymer — (— A —) — 100 — weight — the section — receiving — an end — the alkenyl — a radical — introducing — having had — denaturation — an isobutylene — a system — a block copolymer — (— B —) — 50 – 150 — weight — the section — a plasticizer — (— C —) — zero – 50 — weight — the section — and — a cross linking agent — (— D —) — having blended — a constituent — it is. In this case, to the denaturation isobutylene system block-copolymer (B) 100 weight section in which the alkenyl radical was introduced into the end, a cross linking agent (D) is 0.01 – 20 weight section, and a bridge formation assistant has desirable 0 – 20 weight section. (0038) Moreover, especially the manufacture approach of the thermoplastic-elastomer constituent of this invention is not limited, but if an isobutylene system block copolymer (A), the denaturation isobutylene system block copolymer (B) with which the alkenyl radical was introduced into the end, and the abover-mentioned component used by the case are the approaches which may be mixed by homogeneity, it can adopt all. (0039) Men constructing a bridge dynamically in the denaturation isobutylene system block copolymer (B) with which the alkenyl radical was introduced into the end and manufacturing the thermoplastic-elastomer constituent of this invention at the time of melting mixing of an isobutylene system block copolymer (A) and the denaturation isobutylene system block copolymer (B) with which the alkenyl radical was introduced into the end and manufacturing the thermoplastic-elastomer constituent of this invention at the time of melting mixing of an isobutylene system block co platinum compound, RhCl (PPh3)3, RhCl3, RuCl3, IrCl3, FeCl3, AlCl3, PdCl2 and H2O, NiCl2, and

platinum compound, RhCl (PPh3)3, RhCl3, RuCl3, IrCl3, FeCl3, AlCl3, PdCl2 and H2O, NiCl2, and TiCl4 grade are mentioned. These catalysts may be used independently, and it does not matter even if it uses two or more kinds together. Although there is especially no limit as an amount of catalysts, it is good for using in the range of 10–1 to ten to 8 mol to use in the range of 10–3 to ten to 8 mol to use in the range of 10–3 to ten to 8 mol to use in the range of 10–3 to ten to 8 mol to use in the range of 10–3 to ten to 8 mol to use in the range of 10–3 to ten to 8 mol hardening will not fully advance. Moreover, since the hydrosilylation catalyst is expensive, it is desirable not to use ten – one or more mols. A platinum allyl compound silosane is I among these I the most desirable in respect of compatibility, bridge formation effectiveness, and scorching stability.

[0030] Moreover, for radical crosslinking, it is desirable to make a catalyst share. As a catalyst, radical initiators, such as organic peroxide, are used as a catalyst. It is not limited especially as a radical initiators, such as organic peroxide, are used as a catalyst. It is not limited especially as a radical initiators, such as organic peroxide, as a catalyst. It is not limited especially as a radical initiators, row reample, G t-butyl peroxide, 2, the 5-dimethyl + 2, 5–31 (t-butylperoxy) hexane, 2, the 5-dimethyl + 2, 5–31 (t-butylperoxy) peroxide, the discopropyl peroxide, m-rohloro benzovyl peroxide, 2, 4-dichlorobenzovyl peroxide, diacyl peroxide like discopropyl, Peroxid discopropyl, Peroxide, Peroxyl discoprate [like fault II carbonic acid G 2-ethylhesyl], 1, and 1–31 (t-butylperoxy) - Peroxyl ketal like a 3, 3, and 5-trimethyl cyclohezane etc. can be mentioned. 2, the 5-dimethyl 2, a 5–31-( hexane, 2, the 5-dimethyl 2, and 5-ul-(tert-butylperoxy) hexyne-3 are [ among these ] desirable in respect of odor nature, coloring nature, and scorching stability. [0031] The loadings of organic peroxide are is 8 BUCHIRE at the time of addition of organic peroxide. The range of 0.5 – 5 weight section is desirable to the N system block-copolymer 100

weight section.

[0032] The constituent of this invention can blend the bridge formation assistant which has an ethylene system partial saturation radical on the occasion of the bridge formation processing by organic peroxide. An ethylene system partial saturation radical is a polyfunctional methacrylate monomer like a divinytbenzene, a polyfunctional vinyti monomer like a trianyl SHIANU rate or ethylene glycol dimethacrylate, between glycol dimethacrylate, trientylene glycol dimethacrylate, polyethylene glycol dimethacrylate, trientylene glycol dimethacrylate, and allyl compound methacrylate etc. These may be used independently or may use at least two or more sorts. With such a compound, homogeneity and efficient crosslinking reaction are expectable.

[0033] In order to be easy to deal with ethylene glycol dimethacrylate and triethylene glycol dimethaterylate, to have a peroxide sobublization operation and to work as a distributed assistant (0033) In order to be easy to deal with ethylene gived dimethacrysta and triethylene glycol dimethacrysta, to have a peroxide solubilization operation and to work as a distributed assistant of peroxide also especially in it, since the homogeneity and bridge formation thermoplastic elastomer which was effective and was able to balance hardness and rubber elasticity is obtained, the bridge formation effectiveness by heat treatment is desirable.

[0034] Below 20 weight sections or the addition of the abover-mentioned bridge formation assistant are desirable to the denaturation isobutylene system block-copolymer (B) 100 weight section. Are exceeded, there is an inclination for independent gelation of a bridge formation assistant to tend to progress, and there is a problem in respect of cost. [0035] in the constituent of this invention, since a moldability and flexibility are raised further in addition to an isobutylene system block copolymer (A) and the denaturation isobutylene system block copolymer (B) which has an alkenyl radical at the end, a plasticizer (C) can also be added further, the straight mineral oil used as a plasticizer (C) in the case of processing of rubber — or liquefied or the synthetic softener of low molecular weight can be used. [0036] As straight mineral oil, although the high-boiling point petroleum component of paraffin series, a naphthene, and an aromatic series system is mentioned, the paraffin series and the naphthene which do not check crosslinking reaction are desirable. As liquefied or a synthetic softener of low molecular weight, although there is especially no limit, polybutene, hydrogenation polybutene, liquid polybutadiene, and the loadings of a plasticizer (C) can use one or more sorts. As for the loadings of a plasticizer (C) can use one or more sorts. As for the loadings of a plasticizer (C) can

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a bridge formation catalyst from the middle of the cylinder of an extruder there, and melting kneeding is carried out further. A bridge is dynamically constructed in the denaturation isobutylene system block copolymer (B) with which the alkenyl radical was introduced into the end. The approach of manufacturing the thermoplastic-elastomer constituent which consists of a bridge formation object of the isobutylene system block copolymer (A) of this invention and the denaturation isobutylene system block copolymer (B) with which the alkenyl radical was introduced into the end etc. is employable.

[0042] In performing the above-mentioned method of performing dynamic bridge formation to meking kneading and coincidence, 150–210-degree-C temperature is desirable. Moreover, in this case, an isobutylene system block copolymer (A) cannot cause crosslinking reaction, but can construct a bridge in the denaturation isobutylene system block copolymer (B) with which the alkenyl radical was introduced into the end.

case, an isosoryerie system block copolymer (A) cannot cause crossinning reaction, but can construct a bridge in the denaturation isobutylene system block copolymer (B) with which the alkenyl radical was introduced into the end.

(0043) the denaturation isobutylene system block copolymer with which the alkenyl radical was beforehand introduced into the end — strange — the bridge formation object of (B) is manufactured, and when mixing the bridge formation object with an isobutylene system block copolymer (A) and adjusting the thermoplastic-elastomer constituent of this invention, the approach of illustrating below is adopted preferably.

(0044) To the denaturation isobutylene system block copolymer with which the alkenyl radical was introduced into the above-mentioned end, for example, a cross linking agent and a bridge formation assistant. Add a bridge formation catalyst and it fully kneads at suitable temperature using the kneading machine usually used for manufacture of a rubber bridge formation object. After adopting suitable bridge formation temperature and bridge formation time amount for the bridaned kneading object using the press machine etc. and advancing crosslinking reaction. The bridge formation object of the denaturation isobutylene system block copolymer (B) with which it ground after cooling and the alkenyl radical was introduced into the end can be obtained, and the thermoplastic-elastomer constituent of this invention can be manufactured by carrying out melting mixing of the bridge formation object with an isobutylene system block copolymer (A). (0045) any of the known approach currently conventionally used for manufacture of thermoplastic-elastomer promotentially used for manufacture of thermoplastic or a thermoplastic-elastomer constituent as melting alligation of the bridge formation object of the denaturation isobutylene system block copolymer (B) with which the alkenyl radical was introduced into the end on that occasion, and an isobutylene system block copolymer (B) with which the alkeny

[0047]
[Example] Although this invention is further explained below at a detail based on an example, this invention does not receive a limit at all by these. In addition, in advance of an example, various measuring methods, an appraisal method, and an example are explained.
[0048] (Degree of hardness) Based on JIS K 6325, the test piece used 12.0mm \*\* press sheet.
[0049] (\*\*\*\* breaking strength) Based on JIS K 6251, the test piece pierced and used 2mm thickness press sheet for the No. 3 mold by the dumbbell. The speed of testing was considered as a part for 500mm/.
[0050] (\*\*\*\* elongation after fracture) Based on JIS K 6251, the test piece pierced and used

2mm thickness press sheet for the No. 3 mold by the dumbbell. The speed of testing was considered as a part for 500mm/.

considered as a part for 500mm/. [0051] (Compression set) Based on JIS K 6262, the test piece used 12.0mm thickness press sheet. It measured on condition that deformation 70 degree—Cx 22 hours, and 25%. (Transparency) 2mm thickness press sheet was created, visual observation of the sheet was carried out, that whose background spaces a sheet and can be seen was made into transparence, and what is not visible was made opaque. (Dynamic viscoelasticity) JIS Based on K-6394 (vulcanized nubber and dynamic property test method of thermoplastic rubber), the test piece with a 5mm[ 6mm by ] x thickness of 2mm was out down, and loss tangent tandetts was measured using dynamic viscoelasticity measuring device DVA-200 (IT measurement control company make). The test frequency could be 0.05Hz. Moreover, the cable address and the concrete contents of the ingredient used for below in the example and the example of a comparison are as follows.

example and the example of a comparison are as follows.

SIBS: Polystyrene-polyisobutylene-polystyrene triblock copolymer ASIBS: The denaturation polystyrene-polyisobutylene-polystyrene triblock copolymer with which the alternyl radical was polystyrene-polyisobutyl introduced into the end.

introduced into the end.

IR: Isobutytene isoprene rubber, the product made from JSR (trade name "Butyt065")

Plasticizer: Paraffin series process oil, Idemitsu petrochemical company make (trade name "Diana process oil PW-90")

Cross-finking-agent 1: The chairr-like siloxane cross linking agent 2 which contains an average of five hydrosilyl radicals and an average of five alpha-methyl-styrene radicals in a molecula: A reaction type bromination alkythenol formaldehyde compound, Taoka Chemical Co., Ltd. make (trade name "the tacky roll 250-1")

reaction type orientation asyptement formaterhyde compound, Taoka Chemical Co., Ltd. make (trade name "the tacky roll 250-1")
Bridge-formation assistant 1: Triethylene glycol dimethacrylate, new Nakamura chemistry company make (trade name "NK ester 3G")
Bridge-formation Assistant 2: Zinc-oxide bridge-formation catalyst: 1, 1 and 3 of zerovalent platinum, 3-tetramethyl - 1, 3-diaryl disiloxane complex 1% xylene solution (example 1 of manufacture) [manufacture of a styrene-isobutylene-styrene block copolymer (SIBS)]
A syringe is used after carrying out the nitrogen purge of the inside of the polymerization container of the separable flask of 2L n-hexane (what was dried by molecular sieves) 456.4mL, and butyl chloride (what was dried by molecular sieves) 656.3mL are added. After attaching a polymerization container all over -70-degree C dry ice / methanol bus and cooling, The Siguid-sending tube made from Teffon (trademark) was connected to proof-pressure glass liquefaction extraction tubing with a three-way cook containing isobutylene monomer 232-mL (2871mmol), and the isobutylene monomer was sent by ritrogen pressure in the polymerization container, pJIKUMIRU chloride 0.647g (2.8mmol) and 1.22g (14mmol) of N.N-dimethylacetamide were added. Next, titanium-tetrachloride 8.67mL (79.1mmol) was added further, and the polymerization was started. After agitating at the temperature same for 2.5 hours from polymerization initiation, polymerization solution abbreviation in.ML was sampled as an object for a sampling from the polymerization container. In addition to a lot of water, the reaction was terminated 2 hours the polymerization container. In addition to a lot of water, the reaction was terminated 2 hours after adding this mixed solution.

[0052] The reaction solution was rinsed twice, the solvent was evaporated and the target block

copolymer was obtained by carrying out the vacuum drying of the obtained polymer at 60 degrees C for 24 hours. The molecular weight of the polymer obtained by the gel-perment chromatography (GPC) method was measured. The block copolymer whose Mw of a block copolymer is 101,000 was obtained.

(0053) ((Example 2 of manufacture) Manufacture of the denaturation polystyrene-polyisobutylene-polystyrene triblock copolymer (ASIBS) with which the alkenyl radical was introduced into the end]

A syringe is used after carrying out the nitrogen purge of the inside of the polymerization container of the separable flask of 2L. n-hexane (what was dried by molecular sieves) 480mL, and butyl chloride (what was dried by molecular sieves) 690mL are added. After attaching a

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[0060] (Example 2 of a comparison) SIBS manufactured in the example 1 of manufacture, and IIR at a rate shown in Table 1 Mehing kneading was further carried out at 180 degrees C, and dynamic bridge formation was performed until it carried out netting kneading for 5 minutes using the lab PURASUTO mill (Oriental machine company make) set as 180 degrees C, it added at a rate which, subsequently to Table 1, showed the cross linking agent 2, the bridge formation assistant 3, and the bridge formation assistant 4 and the value of torque showed the peak price (3 - 7 minutes). The obtained thermoplastic-elastomer constituent was able to be easily fabricated at 180 degrees C in the shape of a sheet. The degree of hardness, the \*\*\*\* breaking strength, the \*\*\*\*\* elongation after fracture, and the compression set of the obtained sheet were measured according to the above-mentioned approach. A result is shown in Table 1.

[0061] (Example 3 of a comparison) The constituent was created like the example 1 at a rate shown in Table 1 using ASIBS manufactured in the example 2 of manufacture. However, a sheet-like Plastic solid was not able to be acquired using this constituent.

[0062] (Example 4 of a comparison) The sheet was created using Mitsubishi Chemical RABARON SU5400N, and a degree of hardness, \*\*\*\* breaking strength, \*\*\*\* elongation after fracture and a compression set, transparency, and dynamic viscoelasticity were measured according to the above-mentioned approach. A result is shown in Table 1. [0060] (Example 2 of a comparison) SIBS manufactured in the example 1 of manufacture, and IIR [0063]

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[Table 1]

The thermoplastic-elastomer constituent of this invention has the value of a compression set lower than the SIBS simple substance which is an isobutylene system block copolymer shown in the example 1 of a comparison, and it excels in the compression set, with the property of an isobutylene system block copolymer held. And though a degree of hardness is comparable or soft, it is clear the thermoplastic-elastomer constituent's shown in the bridge formation object shown in the example 2 of a comparison in the example 3.4 as compared with the case where IIR is used to excel in the value of a compression set. Moreover, it is transparent and it is clear that the thermoplastic-elastomer constituent's and an example 1.4 as value of the relation is the short that the thermoplastic elastomer constituent's of an example 1.4 the value of the label in brightness. the thermoplastic-elastomer constituent's of an example 1 the value of tandelta is highly excellent in a vibration-deadening system as compared with the example 4 of a comparis

(0064)
[Effect of the Invention] Thus, a thermoplastic-elastomer constituent is a new thermoplastic elastomer constituent which was rich in flexibility and was excellent in fabrication nature, the rubber-property, the mechanical strength, the compression set property, and the vibration-deadening system, with the property of an isobutylene system block copolymer held. polymerization container all over ~70-degree C dry ice / methanol bus and cooling. The liquid-sending tube made from Teflon (trademark) was connected to proof-pressure glass liquefaction extraction tubing with a three-way cock containing isobutylene monomer 201mL (2132mmol), and the isobutylene monomer was sent by nitrogen pressure in the polymerization tainer, p-JIKUMIRU chloride 2.6g (11.2mmol) and 1.22g (14mmol) of N.N-dimethylacetamide were added. JILLOWAITU Chloride 2.5g (112/mnot) and 1.22g (14/mnot) of N,N-dimethylacetamide were adder Next, titanium-tetrachoride 9.9mt, (90.0mmol) was added further, and the polymerization was started. After agitating at the temperature same for 1.5 hours from polymerization initiation, polymerization solution abbreviation 1mt, was sampled as an object for a sampling from the polymerization solution. Then, styrene monomer 52g (499mmol) was added in the polymerization container. 45 minutes after adding this mixed solution, allyl compound trimethyl silane 12ml (10.0mmol) was added. After stirring at temperature as it is for 60 minutes, a lot of water was added and the reaction was terminated. d and the reaction was terminated.

acced and the reaction was terminated.

(IOS4) The reaction solution was rinsed twice, the solvent was evaporated and the target block copolymer was obtained by carrying out the vacuum drying of the obtained polymer at 50 degrees C for 24 hours. The molecular weight of the polymer obtained by the gel-permeation-chromatography (GPC) method was measured. The block copolymer whose Mw of a block

chromatography (GPC) method was measured. The block copolymer whose Mw of a block copolymer is 22500 was obtained.

[0055] (Example 1) With the rate which showed SIBS manufactured in the example 1 of manufacture of ASIBS manufactured in the example 2 of manufacture in Table 1, melting kneading was carried out for 5 minutes using the lab PURASUTO mill (Oriental machine company make) set as 150 degrees C, and infanticide continuation kneading was added and carried out for minutes at a rate which showed the cross linking agent subsequently to Table 1. The bridge formation catalyst was supplied, melting kneading was carried out further, and dynamic bridge formation was performed. The obtained thermoplastic-elastomer constituent was able to be easily fabricated at 180 degrees C in the shape of a sheet. The degree of Arienass, the \*\*\*\* breaking strength, the \*\*\*\* elongation after fracture and the compression set of the obtained sheet, transparency, and dynamic viscoelasticity were measured according to the above—

breaking strength, the \*\*\*\* elongation after fracture and the compression set of the obtained sheet, transparency, and dynamic viscoelasticity were measured according to the above-mentioned approach. A result is shown in Table 1.

[0056] (Example 2) Except performed dynamic bridge formation as well as an example 1 for the presentation ratio of SIBS and ASIBS. The obtained thermoplastic-elastomer constituent was able to be easily fabricated at 180 degrees C in the shape of a sheet. The degree of hardness, the \*\*\*\* breaking strength, the \*\*\*\* elongation after fracture and the compression set of the obtained sheet, and transparency were measured according to the above-mentioned approach. A standard is element in Table 1. early is shown in Table 1

result is shown in Table 1.

[0057] (Example 3) With the rate which showed SIBS and ASIBS in Table 1, melting kneading was carried out for 5 minutes using the lab PURASUTO mill (Oriental machine company make) set as 150 degrees C, and infanticide continuation kneading was added and carried out for 5 minutes at 150 degrees C, and infanticide continuation kneading was added and carried out for 5 minutes at rate which showed the plasticizer subsequently to Table 1, it added at a rate which whowed the cross linking agent in Table 1, and infanticide continuation kneading was carried out for 5 minutes at the pan. The bridge formation catalyst was supplied, melting kneading was carried out, and dynamic bridge formation was performed. The obtained thermoplastic-elastomer constituent was able to be easily fabricated at 180 degrees C in the shape of a sheet. The degree of hardness of the obtained sheet, a compression set, and transparency were measured according to the abover-mentioned approach. A result is shown in Table 1.

[0058] (Example 4) The sheet was created like the example 3 except having changed the blending ratio of coal. The degree of hardness of the obtained sheet, a compression set, and transparency were measured according to the abover-mentioned approach. A result is shown in Table 1.

Table 1.

[0059] (Example 1 of a comparison) After carrying out melting kneading for 10 minutes using the bp PURASUTO mill which set SIBS manufactured in the example 1 of manufacture as 180 degrees C, it fabricated at 180 degrees C in the shape of a sheet. The degree of hardness, the \*\*\*\* breaking strength, the \*\*\*\* elongation after fracture, the compression set, and transparency of the obtained sheet were measured according to the above-mentioned approach A result is shown in Table 1.

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